UNIVERSITI PUTRA MALAYSIA

EFFECT OF HYDROXYAPATITE ADDITION INTO GLASS IONOMER CEMENT ON PHYSICAL, STRUCTURAL AND MECHANICAL PROPERTIES

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By

WAN NURSHAMIMI BINTI WAN JUSOH

Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfilment of the Requirements for the Degree of Master of Science

January 2021

DEDICATIONS

This thesis is dedicated to:

My parents, Wan Jusoh Wan Ali and Noriza Mohd Yusuf;

My siblings, W. M. Syammil, W. Nursyazwina, W. M. Syazwan, W. M. Syauqi, W. M. Shahmi, W. Nursyafiqa, W. M. Syahir;

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My project co-supervisor, Dr. Mohd Hafiz Mohd Zaid, Dr. Norhazlin Zainuddin;

My teammates, Mohammad Zulhasif Ahmad Khiri, Nadia Asyikin Abdul Rahman, Rohaniah Abdul Jalil;

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My lab mates and friends;

My families;

And others who help me throughout the completion of this thesis.

Thanks for their help, support, understanding, love and encouragement

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Chairman : Associate Professor Khamirul Amin bin Matori, PhD
Faculty : Science

Glass ionomer cement (GIC) is a well-known restorative material applied in dentistry, especially as restorative and luting materials. The present work aims to enhance the physical, structural and mechanical properties of GIC with the addition of hydroxyapatite (HA) since GIC is lacking in the mechanical strength which then limits the use of GIC as restorative material. In this research, waste materials consisting of clam shell (CS) and soda lime silica (SLS) glass are used in the manufacture of alumino-silicate-fluoride (ASF) glass ceramics through melt-quench technique. Meanwhile, synthesized HA powder was obtained by wet chemical precipitation method using CS as the starting material. The control and modified GIC samples were formulated based on a 3:1:1 ratio referring to ASF glass ceramics/HA: polyacrylic acid (PAA): deionized water. All GIC samples were subjected to four different ageing time before being characterized by density measurement, X-ray diffraction (XRD), Fourier transform infrared (FTIR), field emission scanning electron microscopy (FESEM), energy dispersive X-ray (EDX) and compressive strength test. CS and SLS glass are characterized by X-ray fluorescence (XRF) in which the main composition of calcium (Ca) and silicon (Si) respectively encourage the use of waste materials in sample preparation. The existence of fluorapatite (FA) crystal phase in ASF glass ceramics sample was confirmed by XRD, FTIR and FESEM analysis. In addition, the inclusion of HA into the GIC formulation causes an increase in density results. XRD of modified GIC samples detect the presence of fluorohydroxyapatite (FHA) crystal peaks and is confirmed by the OH–F chemical bond at FTIR wavenumber ~3550 cm^{-1}. The morphology of FESEM reveals the formation of spherical particles and agglomerated needle-like belonging to apatite crystals. Moreover, ageing time of control and modified GIC samples did not have a significant effect on the structural properties. The calcium to phosphate (Ca/P) ratio of GIC samples in the range of 1.76 to 3.31

allows the suitability of these materials for implantation purposes. Modified GIC samples show higher compressive strength compared to control GIC. The compressive strength increases with increasing ageing time. GIC added with 5 wt.% of commercial HA at 21 days of ageing time produced the highest compressive strength with 90.12 MPa. Overall, the addition of HA into GIC provides excellent results and better properties to encourage its use as a restorative material in dentistry.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia
sebagai memenuhi keperluan untuk ijazah Master Sains

KESAN PENAMBAHAN HIDROKSIAPATIT KE DALAM SIMEN IONOMER KACA TERHADAP CIRI FIZIKAL, STRUKTUR DAN MEKANIKAL

Oleh

WAN NURSHAMIMI BINTI WAN JUSOH

Januari 2021

Pengerusi : Profesor Madya Khamirul Amin bin Matori, PhD
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Simen ionomer kaca (GIC) adalah bahan pemulihan terkenal yang digunakan dalam pergigian terutamanya sebagai bahan restoratif dan lute. Kerja ini bertujuan untuk meningkatkan sifat fizikal, struktur dan mekanik GIC dengan penambahan hidroksiapatit (HA) kerana GIC kekurangan kekuatan mekanik yang kemudian membatasi penggunaan GIC sebagai bahan pemulihan. Dalam penyelidikan ini, bahan buangan yang terdiri daripada kulit kerang (CS) dan kaca soda kapur silika (SLS) digunakan dalam pembuatan seramik kaca alumino-silicate-fluoride (ASF) melalui teknik lindapan leburan. Sementara itu, serbuk HA yang disintesis diperoleh dengan kaedah pemendakan kimia basah menggunakan CS sebagai bahan permulaan. Sampel GIC kawalan dan diubahsuai dirumuskan berdasarkan nisbah 3:1:1 merujuk kepada seramik kaca ASF/HA: asid poliakrilik (PAA): air deionisasi. Semua sampel GIC dikenakan empat masa penuaan yang berbeza sebelum dicirikan oleh pengukuran ketumpatan, difraksi sinar-X (XRD), inframerah transformasi Fourier (FTIR), mikroskop elektron pengimbas pelepasan medan (FESEM), sinar-X penyebaran tenaga (EDX) dan ujian kekuatan mampatan. CS dan kaca SLS dicirikan oleh pendarfluor sinar-X (XRF) di mana komposisi berat utama kalsium (Ca) dan silikon (Si) masing-masing mendorong penggunaan bahan buangan dalam penyediaan sampel. Kewujudan fasa kristal fluorapatit (FA) dalam sampel seramik kaca ASF disahkan oleh analisis XRD, FTIR dan FESEM. Di samping itu, kemasukan HA ke dalam formulasi GIC menyebabkan peningkatan hasil kepadatan. XRD sampel GIC yang diubahsuai mengesan kehadiran puncak kristal fluorohidroksiapatit (FHA) dan disahkan oleh ikatan kimia OH–F pada nombor gelombang FTIR ~3550 cm^{-1}. Morfologi FESEM memperlihatkan pembentukan zarah sfera dan seperti jarum terkumpul milik kristal apatit. Lebih-lebih lagi, masa penuaan sampel GIC kawalan dan GIC yang diubah tidak memberi kesan yang signifikan terhadap sifat struktur. Nisbah Ca/P sampel GIC dalam julat 1.76 hingga 3.31

membolehkan kesesuaian bahan-bahan ini untuk tujuan implantasi. Sampel GIC yang diubah menunjukkan kekuatan mampatan yang lebih tinggi berbanding dengan GIC kawalan. Kekuatan mampatan meningkat dengan bertambahnya masa penuaan. GIC dengan penambahan 5 wt.% HA komersial pada 21 hari masa penuaan menghasilkan kekuatan mampatan tertinggi dengan 90.12 MPa. Secara keseluruhan, sampel GIC yang ditambah dengan HA memberikan hasil yang sangat baik dan sifat yang lebih baik untuk mendorong penggunaannya sebagai bahan pemulihan dalam pergigian.

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This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfilment of the requirement for the degree of Master of Science. The members of the Supervisory Committee were as follows:

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LIST OF ABBREVIATIONS AND SYMBOLS

HA	Hydroxyapatite
GIC	Glass ionomer cement
CS	Clam shell
SLS	Soda lime silica
ASF	Alumino-silicate-fluoride
PAA	Polyacrylic acid
SBF	Simulated body fluid
HCA	Hydroxycarbonate apatite
nCDHA	nanocrystalline calcium deficient hydroxyapatite
FA	Fluorapatite
FHA	Fluorohydroxyapatite
XRF	X-ray fluorescence
EDX	Energy dispersive X-ray
XRD	X-ray diffraction
FESEM	Field emission scanning electron microscopy
FTIR	Fourier transform infrared spectroscopy
TGA	Thermogravimetric analysis
Ca/P	Calcium to phosphate
P/L	Powder to liquid
ICDD	International Center Diffraction Data
n	Integer
λ	Wavelength
d_{plane}	Distance between the lattice planes
θ	Angle between the incident and lattice plane

ρ	Density
m	Mass
V	Volume
ρ_{sample}	Density of sample
W_{air}	Weight of sample in air
$W_{distilled\ water}$	Weight of sample in distilled water
$\rho_{distilled\ water}$	Density of distilled water
wt.%	Weight percentage
σ	Compressive strength
F	Maximum load applied
d_{sample}	Average diameter of sample

CHAPTER 1

INTRODUCTION

This chapter presents a concise description about the background of study regarding the effect of hydroxyapatite (HA) addition into glass ionomer cement (GIC). Besides, problem statements and research aims are also stated in this chapter. This is accompanied by importance of the study and also thesis outline as a framework which is significant to this study.

1.1 Research background

The application of biomaterials and bioceramics in the medical and dental field is being extensively studied by researchers. Materials which are implanted and used in the body are known as biomaterials. Biomaterials can be divided into two categories, known as natural and synthetic biomaterials. Natural biomaterials are any materials produced from plants or animals and used to replace or restore impaired body tissues and organs. (El-Meliegy and Noort, 2012; Parida et al., 2012). According to Basu and Nath (2010), earliest use of natural biomaterials was reported from ancient Egypt, whereby people created sutures from animal sinew. Meanwhile, materials that were created and engineered with the purpose of implantation in the human body are called synthetic biomaterials. Generally, synthetic biomaterials are classified into metals, polymers, ceramics and semiconductor materials (El-Meliegy and Noort, 2012). The classification of synthetic biomaterials is depicted in Figure 1.1.

In the field of dentistry, the application of biomaterials is also a concern where the use of materials that respond well and are safe to use in the human oral environment, especially tooth anatomy can help improve tooth structure as well as their function. For dental implant, the earliest reported materials applied in human dentistry are amalgam, composite resins and cements (Khoroushi and Keshani, 2013). Amalgam, the most popular researched filling material in dental treatment has been used for more than a century. The main use of amalgam or known as silver fillings in dental treatment is to fill cavities in tooth structure caused by tooth decay. Basically, amalgam is formed by mixing metals which include mercury and alloy powder composed of silver, tin and copper. This reaction resulted in amalgam production with high mechanical strength and excellent durability. However, lack of aesthetics properties, low thermal sensitivity and the mercury content in amalgam composition create a controversy on the toxicity and allergy to the human body, thus restricting the use of amalgam as dental filling.

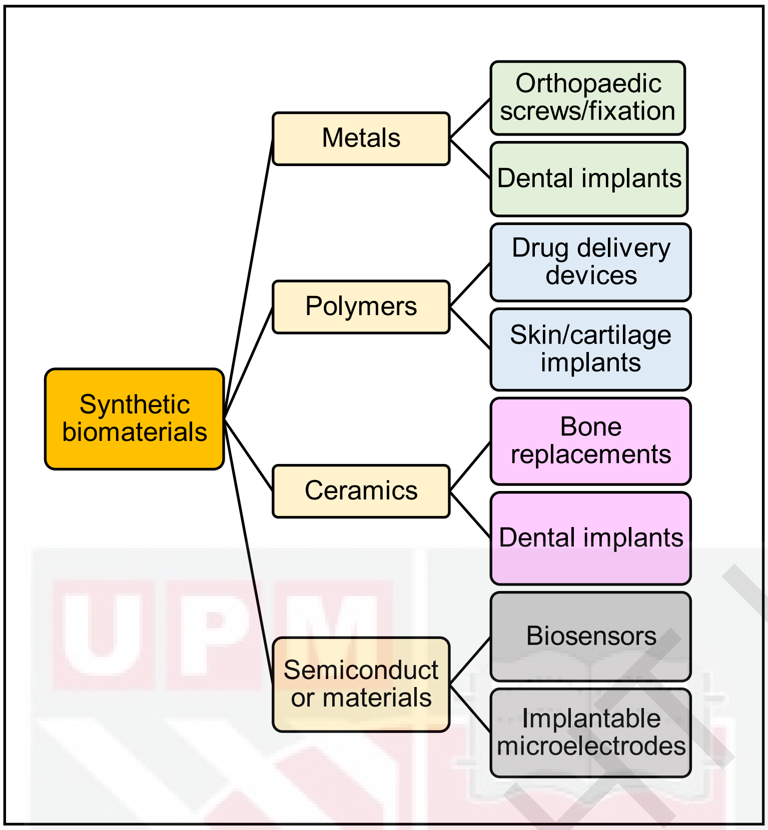

Figure 1.1 : Classification of synthetic biomaterials
(El-Meliegy and Noort, 2012)

Therefore, various types of dental products had been created along the needs to fulfil the requirement of a dental implant especially for repairing and restoration of tooth structure. Glass ionomer cement (GIC), also recognized as glass polyalkenoate cement, is a biomaterial that was introduced as a luting and restorative material in dental applications. Wilson and Kent discovered GIC in the early of 1970s (Wilson and Kent, 1972). This well-known restorative material composed of powdered glass which reacted in acid base reaction with the existence of water as a medium of reaction. According to Akinmade and Nicholson (1993), GIC is a water-based cement in which the glass powder and polyalkenoic acid experience acid base reaction upon combining.

One of the noticeable applications of dental cements is to secure metal and non-metal inlays, crowns and bridges to the surface of the dentures permanently for the purpose of restoring tooth's appearance, structure or function. GIC is one of the dental fillings materials which are available in dentistry. The use of GIC can be more advantageous especially for physical appearance since it is known as tooth-colored material. Besides, fluoride ion release from the GIC system makes it to be considered as therapeutic dental materials, whereby the fluoride ion is important for the remineralization process in preventing tooth decay.

Due to outstanding properties such as biocompatible with the tissues in tooth structure, have good adhesion properties, stable in aqueous environment and lack of exothermic polymerization, the use of GIC in dental application is a concern. Other than that, fluoride release from GIC especially in acidic conditions helps in preventing tooth decay since fluoride can resist the demineralization process. Some research measured into the ideal conditions for a material to function as a dental cement in terms of biological, chemical, thermal, mechanical, and other influences. (Sita et al., 2014; Sidhu and Nicholson, 2016). However, instead of having excellent properties as restorative material, the use of GIC is still limited since it performs poor mechanical properties. Therefore, in order to overcome these drawbacks, several researches were carried through and aimed at upgrading the mechanical characteristics of GIC. One of the attempts made to solve the problem was the inclusion of ceramics material or bioceramics.

Hydroxyapatite (HA) is a well-known bioceramics which belongs to calcium phosphate group with calcium to phosphate (Ca/P) ratio 1.67 and $Ca_{10}(PO_4)_6(OH)_2$ as molecular formula, close to the bone and tooth structure apatite composition (Arita et al., 2011; Bardhan et al., 2011; Goenka et al., 2012; Kantharia et al., 2014; Pepla et al., 2014). HA promises toughness comparable to natural bone and tooth along with excellent biocompatibility (Wang et al., 2010; Khiri et al., 2016). Enamel, dentin and cementum which are parts of tooth structure contain high composition of HA which is responsible for biocompatibility and osteoconductivity of the tooth (Pepla et al., 2014; Pajor et al., 2019). Furthermore, HA exhibits properties such as non-toxicity, non-immunogenic, non-inflammatory and good bioactivity, making it an excellent material for clinical use, especially for bone scaffolds and dental implant materials (Sadat-Shojai et al., 2013; Rujitanapanich et al., 2014).

In this study, the formulation of GIC is based on the mixing of alumino-silicate-fluoride (ASF) glass ceramics as base silicate powder, polyacrylic acid (PAA) as polyacid and water as a reaction medium. The utilization of waste materials such as clam shell (CS) and soda lime silica (SLS) glass in the fabrication of ASF glass ceramics and also CS in the synthesize of synthesized HA powder, being an interesting subject to research. Based on Awang-Hazmi et al. (2007), major mineral composed in CS is calcium carbonate ($CaCO_3$) at about 98.70% which then can be converted to calcium oxide (CaO) by calcination process (Awang-Hazmi et al., 2007; Jusoh et al., 2019; Rahman et al., 2019). Meanwhile, silicon dioxide (SiO_2) is the main composition of the SLS glass which is composed about 73.9 % of the silica (Thoo et al., 2013). Thus, CS and SLS glass are used as replacement for the respective CaO and SiO_2 sources. Moreover, the utilization of waste materials can help in reducing the environmental pollution and help in producing the economical paste cements.

1.2 Problem statements

Waste materials which are CS and SLS glass are utilized in the sample preparation as CaO and SiO_2 sources, respectively. The use of these waste materials can be a great help in reducing the disposal problem and also reduce the cost of material fabrication.

GIC is a very beneficial restorative material used in dentistry since it is biocompatible and performs good chemical adherence to tooth structure (Rahman et al., 2017; Khiri et al., 2020). Besides, fluoride ion release performed by GIC after being applied to human teeth is important in preventing tooth decay. However, despite having many advantages, GIC is lacking in terms of mechanical properties. Weak mechanical properties have been established including poor flexural strength, low fracture strength and toughness, weak wear resistance as well as opaqueness, thereby restricting the use of GIC as restorative material in dentistry (Lohbauer, 2010).

Researchers have attempted a variety of methods to develop mechanical properties of GIC, including adding other fillers to strengthen it. For example, reinforcement with metal powders (Irie and Nakai, 1988), modification with resin (Farrugia and Camilleri, 2015), incorporation with SiC whiskers/short fibers (Arita et al., 1992; Kobayashi et al., 2000; Lihua et al., 2010) as well as HA (Khaghani et al., 2016a; Alatawi et al., 2019), fluorapatite (FA) nanobioceramics (Moshaverinia et al., 2008) and forsterite nanoparticles (Sayyedan et al., 2014). The inclusion of HA powder into the GIC composition is aimed at enhancing the mechanical, physical, and structural properties of the resulting GIC in this research.

The mechanical strength of the resulting GIC will be improved by the presence of HA crystal. This is because an intermediate layer forms between the HA crystal and the GIC matrix, which is highly resistant to acid attack and difficult to break (Moshaverinia et al., 2008; Arita et al., 2011). Excessive amounts of HA in GIC, conversely, will reduce mechanical strength of the resulting GIC because the chemical bonding between the HA particles and the polymeric chain of the GIC matrix weakens (Khaghani et al., 2016a). Therefore, the investigation on the suitable ratio of HA to be incorporated with ASF glass ceramics powder for GIC production is important to find the suitable amount of HA addition for optimum effects especially for mechanical properties.

1.3 Objectives of study

This research concentrated on the addition of HA into GIC in contemplation of improving the mechanical properties of resulting GIC. Therefore, the principal aims of this research are:

1. To prepare ASF glass ceramics powder derived from CS and SLS glass.
2. To study the effect of commercial and synthesized HA into GIC composition on physical, structural and mechanical properties of the modified GIC
3. To investigate the suitable ratio of commercial and synthesized HA to be incorporated with ASF glass ceramics powder for GIC production.
4. To examine the effect of ageing time on control and modified GIC samples.

1.4 Importance of study

The research of HA added to GIC is important in order to improve the modified GIC's physical, structural, and mechanical properties, which are generally used in dentistry. Besides, the investigation of suitable ratio of HA to be added with ASF glass ceramics powder before being formulated to GIC is conducted by analyzing the optimum results of the modified GIC. Since there is limited studies of formulation of GIC from waste, this can lead to extensive research of formulating GIC by using waste materials and the improvement of the properties of the resulting GIC.

1.5 Outline of thesis

The description of this thesis is divided into five chapters. Chapter 1 gives an explanation on the background of study, problem statements, objectives and importance of study. Chapter 2 covers the previous works that had been done by other researchers and mostly focused on the biomaterials and bioceramics, GIC, HA, ASF glass ceramics and HA addition into GIC composition. Meanwhile, Chapter 3 focuses on the methodology used in this work starting from preparation of raw materials, fabrication of ASF glass ceramics powder, HA powder, GIC samples and also sample characterization. Next, the results concerning the effect of incorporating HA into GIC on their physical, structural and mechanical properties are analyzed and discussed in Chapter 4. Lastly, Chapter 5 gives the conclusion and also the recommendation for future works.

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